



## INTERNATIONAL APPLICATION PUBLISHED UNDER THE PATENT COOPERATION TREATY (PCT)

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<b>(21) International Application Number:</b> PCT/US99/24795 <b>(22) International Filing Date:</b> 22 October 1999 (22.10.99) <b>(30) Priority Data:</b> 09/186,537 5 November 1998 (05.11.98) US <b>(71) Applicant:</b> SIKA CORPORATION [US/US]; 30800 Stephenson Highway, Madison Heights, MI 48071 (US). <b>(72) Inventors:</b> CHANG, Chin-Jui; 6598 Hilltop Drive, Troy, MI 48098 (US). FITZGERALD, Gerald; 38737 Golfview, Clinton Township, MI 48038 (US). <b>(74) Agent:</b> VAN HOOZER, Thomas, H.; Hovey, Williams, Timmons & Collins, Suite 400, 2405 Grand Boulevard, Kansas City, MO 64108 (US).		<b>(81) Designated States:</b> AL, AM, AT, AU, AZ, BA, BB, BG, BR, BY, CA, CH, CN, CU, CZ, DE, DK, EE, ES, FI, GB, GD, GE, GH, GM, HR, HU, ID, IL, IN, IS, JP, KE, KG, KP, KR, KZ, LC, LK, LR, LS, LT, LU, LV, MD, MG, MK, MN, MW, MX, NO, NZ, PL, PT, RO, RU, SD, SE, SG, SI, SK, SL, TJ, TM, TR, TT, UA, UG, UZ, VN, YU, ZW, ARIPO patent (GH, GM, KE, LS, MW, SD, SL, SZ, TZ, UG, ZW), Eurasian patent (AM, AZ, BY, KG, KZ, MD, RU, TJ, TM), European patent (AT, BE, CH, CY, DE, DK, ES, FI, FR, GB, GR, IE, IT, LU, MC, NL, PT, SE), OAPI patent (BF, BJ, CF, CG, CI, CM, GA, GN, GW, ML, MR, NE, SN, TD, TG).  <b>Published</b> <i>With international search report.</i>
<b>(54) Title:</b> SOUND DEADENING AND STRUCTURAL REINFORCEMENT COMPOSITIONS AND METHODS OF USING THE SAME		
<p>The diagram shows a U-shaped member 10. It consists of a base portion 20 and two leg portions 22 and 24. The base portion 20 has a base section 34. The leg portions 22 and 24 have leg sections 36 and 38 respectively. The member 10 is shown in a perspective view, with dashed lines indicating internal structure or alternative configurations.</p>		
<b>(57) Abstract</b> <p>Expandable sealant and baffle compositions and methods of forming and using such compositions are provided. The compositions comprise a first thermoplastic resin, an epoxy resin, preferably a different second thermoplastic resin, and optionally a compound selected from the group of pigments, blowing agents, catalysts, curing agents, reinforcers or mixtures thereof. The compositions are heat-expanded into lightweight, high strength bodies for sealing hollow structural members of vehicles and decrease the noise of the members. In a preferred embodiment, the first thermoplastic resin is an SBS block co-polymer, the epoxy resin is a bisphenol-A based liquid epoxy resin, the second thermoplastic resin is a polystyrene, and the reinforcer is hydrated amorphous silica. The compositions are formed on apparatus (10) which comprises a U-shaped member (12) which comprises leg portions (22 and 24), base portion (20), base section (34) and leg sections (36 and 38) all of which can be supported on lattice-type nylon supports.</p>		

## SOUND DEADENING AND STRUCTURAL REINFORCEMENT COMPOSITIONS AND METHODS OF USING THE SAME

### BACKGROUND OF THE INVENTION

#### 5 Field of the Invention.

The present invention is broadly concerned with expandable sealant and baffle compositions for sealing hollow structural members of vehicles, and methods for making and using such compositions. The compositions are prepared by forming an expandable mixture including a first thermoplastic resin (preferably an SBS block co-  
10 polimer) and an epoxy resin (preferably a bisphenol A-based liquid epoxy resin). The compositions preferably also include a second thermoplastic resin (preferably a polystyrene) different from the first thermoplastic resin, and a compound selected from the group consisting of pigments, blowing agents, catalysts, curing agents, reinforcers, and mixtures thereof. The preferred reinforcers are hydrated amorphous silica and glass  
15 microspheres. The compositions of the invention are injection moldable and can be formed into freestanding, self-sustaining parts. Alternately, the compositions of the invention can be supported on lattice-type nylon supports. Upon heating of the compositions to temperatures of at least about 300°F, the compositions greatly expand to form lightweight products having high compressive strengths.

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#### Description of the Prior Art.

During the fabrication of automobiles, trucks, and similar over-the-road vehicles, many body components present structural members having cavities that require sealing to prevent the entrance of moisture and contaminants which can cause  
25 corrosion of the body parts. It is also desirable to greatly strengthen the members while maintaining their light weight. It is also necessary to stabilize these members in order to attenuate noise that would otherwise be transmitted along the length or passage of the cavity. Many of these cavities are irregular in shape or narrow in size, thus making them difficult to properly seal and baffle.

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Many attempts have been made to seal these cavities, spraying sealants into the cavity, introducing foam products into the cavity, and using of fiberglass matting and the like. These methods each have their drawbacks. For example, foaming in place presents a problem in that it is difficult to control where the foam travels upon its introduction into the cavity. Furthermore, it is often necessary to introduce an excess  
35 amount of foam into the cavity in order to ensure that the cavity is sufficiently sealed. Finally, foams will generally not adhere to the interior surfaces of the cavity walls if those surfaces contain even a small amount of oil.

the length of the cavity. Furthermore, this composition should have a high compressive strength so that it reinforces the structural members in which it is used.

### SUMMARY OF THE INVENTION

5 The instant invention overcomes these problems by providing an expandable sealant and baffle compositions comprising mixtures of thermoplastic resin(s) and an epoxy resin which are injection moldable and lightweight, and which have a high compressive strengths.

10 In more detail, the compositions of the invention include a first thermoplastic resin, usually selected from the group consisting of the polystyrenes, rubbers (preferably solid rubbers), and mixtures thereof. It is preferred that the first thermoplastic resin be a solid rubber or mixtures of solid rubbers. Preferred solid rubbers include styrene-butadiene rubber (such as SBR 1009<sup>®</sup>), nitrile-butadiene rubber (such as Nipol 1411<sup>®</sup>), thermoplastic elastomers including SBS block co-polymers (such as Fina Clear 530<sup>®</sup>), and mixtures thereof. (As used herein, "rubber" is intended to include all synthetic rubbers as well as elastomers.). If the rubber used is an SBS block co-polymer, it is preferred that the SBS block co-polymer have a molecular weight of from about 100,000-150,000, and preferably from about 110,000-135,000. When a styrene-butadiene rubber is used as the rubber, the ratio of butadiene to styrene is preferably from about 32:68 to about 18:82, and more preferably from about 27:73 to about 23:77. If the first thermoplastic resin is a rubber, then the rubber is preferably present in the composition at a level from about 20-30% by weight, and more preferably from about 23-28% by weight, based upon the total weight of the composition taken as 100% by weight.

25 If the first thermoplastic resin is a polystyrene, then the polystyrene should be present in the composition at a level of from about 5-20% by weight, and preferably at a level of from about 10-15% by weight, based upon the total weight of the composition taken as 100% by weight. It is preferable that the polystyrene have a molecular weight of from about 150,000-320,000, and more preferably from about 200,000-270,000. Two preferred polystyrenes are sold under the trade names Fina Crystal 500<sup>®</sup> and Fina Crystal 535<sup>®</sup>.

30 The compositions further include an epoxy resin, preferably a liquid epoxy resin such as a bisphenol A-based liquid epoxy resin. The epoxy resin should be present in the composition at a level of from about 30-45% by weight, and preferably from about 35-40% by weight, based upon the total weight of the composition taken as 100% by weight. A preferred solid epoxy resin is available under the trade name Epon 1001 F<sup>®</sup>.

Table 1

Compound	% By Wt. <sup>a</sup>	Examples
Pigments	0-5%	Carbon black, zinc oxide
Blowing agents	0-5%	Azodicarbonamides such as the following which are available from Uniroyal - Celogen AZ 765 <sup>®</sup> , Celogen AZ 754A <sup>®</sup> , and Celogen AZ 130 <sup>®</sup>
Catalysts	0-5%	Urea-based catalysts such as N,N dimethyl phenyl urea and catalysts sold under the trade names MBTS <sup>®</sup> , Dicap <sup>®</sup> , and Urisys U 405 <sup>®</sup>
Curing agent	0-5%	Sulfur, dicyandiamide, including the curing agent sold under the trade name Urisys DDA 10 <sup>®</sup>

<sup>a</sup> Percent by weight of the particular compound, based upon the total weight of the composition taken as 100% by weight.

The preferred reinforcers are selected from the group consisting of hydrated amorphous silica, glass microspheres, and mixtures thereof. Preferably the compositions hereof include from about 1-10% by weight hydrated amorphous silica and from about 10-20% glass microspheres. Utilizing both of these reinforcers results in a composition having a very high compressive strength. Also, hydrated amorphous silica is important for providing a composition that has high expansion capabilities as well as a viscosity (at 110°C) of less than about 1500 P, and preferably less than about 1250 P, so that the composition can readily be injection molded.

One of the most preferred embodiments of the compositions in accordance with the instant invention includes the following: from about 20-30% by weight SBS block co-polymer; from about 5-20% by weight polystyrene; from about 30-45% by weight bisphenol A-based liquid epoxy resin; from about 0.5-5% by weight carbon black; from about 1-10% by weight hydrated amorphous silica; from about 10-20% by weight glass microspheres; from about 0.5-5% by weight blowing agent; from about 0.3-5% by weight catalysts; and from about 1-5% by weight curing agent, with all percents by weight being based upon the total weight of the composition taken as 100% by weight.

The compositions of the invention are formed by mixing the first thermoplastic resin (preferably solid rubber) with a small portion (about 1/40th of the total amount) of the epoxy resin in a heated mixer until the temperature of the mixer reaches from about 240-260°F (the temperature of the mixture within the mixer is at least about 175°F) and the mixture is substantially homogeneous, at which time all of the second thermoplastic resin (preferably a polystyrene) is added to the mixer and mixing is continued. After the second thermoplastic resin is substantially mixed with the first

structure is superior to the structure in U.S. Patent No. 5,755,486 to Wycech as the use of a lattice support element adds minimal weight to the apparatus.

The expanded composition then adheres to adjacent walls of the cavity in which the apparatus is placed. Preferred apparatuses comprise a plurality of U-shaped expandable composition structures spaced apart from one another and in general alignment, having the lattice support element attached to the three outer surfaces of the U-shaped structure. This embodiment provides for contact by the composition with a greater surface area of the cavity walls than is provided by U.S. Patent No. 5,506,025 to Otto et al., thus providing superior reinforcement of structural member forming the cavity.

In use, the compositions of the invention are formed into the U-shaped structure described above, or are shaped (in either free standing form or some type of lattice-supported form) to conform to the cross-sectional shape of (although slightly smaller than) the cavity in which the composition or apparatus is to be used. The formed composition or apparatus is then placed within the hollow channel or cavity and heat is applied so that the composition expands as the epoxy resin is crosslinked and the expanded composition adheres to the sidewalls of the cavity resulting in a channel or cavity that is substantially blocked by the expanded composition. Optionally, some type of fastening device or mechanism can be utilized to secure the composition within the channel prior to heat expansion.

It will be appreciated that the compositions of the invention can be used in virtually any area of the automotive industry, including body shops, paint shops, and automobile manufacturing facilities. A particular advantage of the compositions and apparatuses of the invention is that they can be placed within the desired channel prior to conveyance of the vehicle body through a bake oven where the temperatures are sufficiently high to expand the composition.

When the sealants and baffle compositions of the invention are subjected to a temperature of at least about 300°F, the percent expansion of the composition will be at least about 95%, preferably at least about 125%, and more preferably at least about 150%, wherein the percent expansion (as used herein) is defined as:

$$100 \times \{[(\text{the specific gravity of the composition before heating}) - (\text{the specific gravity of the composition after heating})]/(\text{the specific gravity of the composition after heating})\}.$$

The expanded compositions have a compressive strength (using a sample having a diameter of 2 inches and a length of 4 inches and a compression rate of 0.5 inches/minute) of at least about 1200 psi, preferably at least about 1400 psi, and more preferably at least about 1600 psi. Prior to expansion, the compositions have a specific

These depressions correspond in both size and shape with the size and shape of the latticework of element 18. The depressions are preferably at least as deep as the lattice is thick, and act as a retaining mechanism so that element 18 remains attached to members 12, 14, 16. Thus, the expandable composition of which members 12, 14, 16 are formed protrudes somewhat through the lattice openings of element 18 (as indicated by numeral 42 in Figs. 2-4). Element 18 can be formed so that each of the sections 34, 36, 38 are integral, or alternately, each of sections 34, 36, 38 can be formed separately and connected by some fastening mechanism (such as clips or snaps) around the members 12, 14, 16 of the apparatus.

In operation, the apparatus 10 is positioned within the channel of a hollow member (such as within a vehicle). The apparatus 10 can be positioned at any angle and orientation as determined appropriate by one having ordinary skill in the art. For example, the base portions 20, 30, 32 and base section 34 of apparatus 10 can be positioned against the lower wall of the channel, while the respective leg portions and sections of apparatus 10 would be positioned against the sidewalls of the channel. Or, the apparatus 10 could be inverted so that the base portions 20, 30, 32 and base section 34 are positioned against the upper wall of the channel.

Once the apparatus 10 is positioned within the channel, the apparatus 10 is then subjected to heat (such as by passing the vehicle through a bake oven), preferably having a temperature of at least about 300°F. The heat application will cause the composition to expand in all directions, thus protruding even further through the openings of the latticework. The expanded composition will then adhere to the walls of the channel, securing apparatus 10 within the channel.

While the figures illustrate an embodiment utilizing a support structure for the compositions of the invention, those skilled in the art will understand that the compositions of the invention do not require a support structure and can be used in a freestanding, self-sustaining form. Also, numerous other support elements can be used with the compositions of the invention depending upon the particular application.

## EXAMPLES

The following examples set forth preferred compositions and methods in accordance with the invention. It is to be understood, however, that these examples are provided by way of illustration and nothing therein should be taken as a limitation upon the overall scope of the invention.

### EXAMPLE 1

#### 1. Preparation of Premix

The pellets were added to the hopper (not equipped with a stirrer) of a conventional injection mold machine and molded into the desired shapes at about 190°F.

5           4.   Test Properties of the Molded Product

The mechanical properties of the compound were tested. Those properties are reported in Table 2 below. These tests were carried out as follows on 10 mm thick, 1 in. x 1 in. portions of the product prepared as previously described:

- 10           (1)   Specific Gravity Before Bake.
- (2)   Specific Gravity After Bake - The composition was placed in an oven heated to 325°F. The composition was kept in the oven for 10 minutes after the temperature of the composition reached 325°F (referred to as 325°F for 10 minutes Metal Temperature).  
15           The composition was then removed from the oven and allowed to remain at room temperature (RT) for 60 minutes (referred to as RT for 60 minutes). This was followed by 325°F for 10 minutes Metal Temperature, then RT for 60 minutes, and finally 250°F for 20 minutes Metal Temperature, followed by the  
20           determination of the specific gravity.
- (3)   Expansion After Bake = [(specific gravity before bake) - (specific gravity after bake)]/(specific gravity after bake)
- (4)   Compressive Strength After Bake - The compressive strength was determined after the following was carried out: the  
25           composition remain in a 350°F oven for a total of 35 minutes; RT for 60 minutes; and finally 250°F for 20 minutes metal temperature.

## EXAMPLE 3

A molded compound was formed following the procedures set forth in Example 1 except the following components and concentrations were used: 120 grams of Fina Crystal 500<sup>®</sup>; 240 grams of Fina Clear 530<sup>®</sup>; 40 grams of Nipol 1411<sup>®</sup> (nitrile-butadiene rubber); 400 grams of Araldite 6010<sup>®</sup>; 10 grams of carbon black; 4.8 grams of zinc oxide; 45 grams of HiSil 233<sup>®</sup>; 155 grams of Scotchlite S60/10,000<sup>®</sup>; 30 grams of Urisys DDA 10<sup>®</sup>; 4 grams of Urisys U 405<sup>®</sup>; and 6 grams of Celogen AZ 130<sup>®</sup>. The final product was tested for its mechanical properties as set forth in Table 4.

Table 4

MECHANICAL TEST	RESULTS
Specific Gravity Before Bake	0.9
Specific Gravity After Bake	0.39
Percent Expansion	131
Compressive Strength, psi <sup>a</sup>	1822 <sup>b</sup>

<sup>a</sup> The test specimens were aged in a 90°C oven for four hours and tested immediately upon removal from the oven.

<sup>b</sup> Several tests of the compressive strength were conducted on this sample. This number is the average of the results of all tests conducted.

## EXAMPLE 4

Several more compositions were prepared following the preparation procedures set forth in the preceding examples. The formulations and mechanical properties of the resulting compounds are set forth in Tables 5 and 6, respectively.



the composition was as described in the previous examples. The components and concentrations used were as follows: 133.33 grams of Fina Crystal 500<sup>®</sup>; 266.67 grams of Fina Clear 530<sup>®</sup>; 400 grams of Araldite 6010<sup>®</sup>; 10 grams of carbon black; 4.8 grams of zinc oxide; 45 grams of Cabot TS-720<sup>®</sup> (fumed silica); 155 grams of Scotchlite S60/10,000<sup>®</sup>; 30 grams of Urisys DDA 10<sup>®</sup>; 4 grams of Urisys U 405<sup>®</sup>; and 6 grams of Celogen AZ 130<sup>®</sup>. The properties of the resulting compound are set forth in Table 7 below.

Table 7

MECHANICAL TEST	RESULTS
Specific Gravity Before Bake	0.93
Specific Gravity After Bake	0.43
Percent Expansion	116
Compressive Strength, psi <sup>a</sup>	1572 <sup>b</sup>
Viscosity (110°C), P	3600

<sup>a</sup> The test specimens were aged in a 90°C oven for four hours and tested immediately upon removal from the oven.

<sup>b</sup> Several tests of the compressive strength were conducted on this sample. This number is the average of the results of all tests conducted.

In comparing these results to the results of the previous examples, it can be seen that this compound has a lower expansion than the other compounds. Furthermore, the viscosity is much higher than the viscosity of the compound obtained in Example 2, where hydrated amorphous silica was used. With such a high viscosity, this compound would not be useful in injection molding.

#### EXAMPLE 6

This test was carried out to determine the effect on the mechanical properties of the composition if no polystyrene was used. The procedure followed to prepare the composition was as described in the previous examples. The components and concentrations used were as listed in Example 2, with the exception that no Fina Crystal 500<sup>®</sup> (or any other polystyrene) was used. The properties of the resulting compound are set forth in Table 8 below.

Table 9

MECHANICAL TEST	RESULTS
Specific Gravity Before Bake	0.91
Specific Gravity After Bake	0.35
Percent Expansion	160
Compressive Strength, psi <sup>a</sup>	1095 <sup>b</sup>

<sup>a</sup> The test specimens were aged in a 90°C oven for four hours and tested immediately upon removal from the oven.

<sup>b</sup> Several tests of the compressive strength were conducted on this sample. This number is the average of the results of all tests conducted.

10. The body of claim 4, wherein said first thermoplastic resin is an SBS block co-polymer and is present in said composition at a level of from about 20-30% by weight, based upon the total weight of the composition taken as 100% by weight.

5 11. The body of claim 1, further including a quantity of a second thermoplastic resin different from said first thermoplastic resin and selected from the group consisting of the polystyrenes, solid rubbers, and mixtures thereof.

10 12. The body of claim 11, wherein the total weight percent of said first thermoplastic resin and said second thermoplastic resin is from about 25-50% by weight, based upon the total weight of the composition taken as 100% by weight.

15 13. The body of claim 11, further including a quantity of a third thermoplastic resin different from said first and second thermoplastic resins and selected from the group consisting of polystyrenes, solid rubbers, and mixtures thereof.

14. The body of claim 1, wherein said epoxy resin is a liquid epoxy resin.

20 15. The body of claim 14, wherein said liquid epoxy resin is a bisphenol A-based liquid epoxy resin.

16. The body of claim 1, wherein the percent expansion of said body is at least about 125%.

25 17. The body of claim 1, wherein said body is expanded by heating to a temperature of at least about 300°F and said expanded body has a compressive strength of at least about 1200 psi.

30 18. The body of claim 1, wherein said body is expanded by heating to a temperature of at least about 300°F and said expanded body has a specific gravity with reference to water of less than about 0.47.

35 19. The body of claim 1, wherein said body has a specific gravity with reference to water of at least about 0.90.

20. The body of claim 1, wherein said body is formed by injection molding.

28. An injection moldable, reinforcing sealant and baffle body comprising an expanded, self-sustaining synthetic resin composition, said composition comprising:  
a quantity of a first thermoplastic resin;  
a quantity of a second thermoplastic resin different from said first thermoplastic resin; and  
a quantity of an epoxy resin,  
said first and second resins and said epoxy resin being reacted together,  
said body having a compressive strength of at least about 1200 psi and a specific gravity with reference to water of less than about 0.47.

29. The body of claim 28, wherein said first thermoplastic resin is a polystyrene, said second thermoplastic resin is an SBS block co-polymer, and said epoxy resin is a bisphenol A-based liquid epoxy resin.

30. The body of claim 29, including from about 30-45% by weight of a bisphenol A-based liquid epoxy resin, from about 5-20% by weight of a polystyrene, and from about 20-30% by weight of an SBS block co-polymer, based upon the total weight of the composition taken as 100% by weight.

31. The body of claim 30, further including a compound selected from the group consisting of pigments, blowing agents, catalysts, curing agents, reinforcers, and mixtures thereof.

32. The body of claim 31, wherein said composition includes from about 1-10% by weight hydrated amorphous silica, based upon the total weight of the composition taken as 100% by weight.

33. The body of claim 32, wherein said composition further includes from about 10-20% by weight glass microspheres, based upon the total weight of the composition taken as 100% by weight.

34. The body of claim 33, wherein said body has a compressive strength:specific gravity after bake ratio of at least about 2500:1.

42. The method of claim 41, wherein said process for forming the pre-mix further includes the step of mixing a second thermoplastic resin different from said first thermoplastic resin with the mixture of step (a).

5 43. The method of claim 41, further including the step of forming the intermediate mixture resulting from step (b) into pellets prior to step (c).

44. The method of claim 41, further including the step of shaping the product resulting from step (c) to conform to the shape of an opening defined by a structural member to be sealed by said product.  
10

45. The method of claim 44, further including the step of placing said shaped product into said opening within said structural member.

15 46. The method of claim 45, further including the step of heating said structural member containing said shaped product by subjecting said member to air having a temperature of at least about 300°F to expand said shaped product.

20 47. The method of claim 46, wherein said expanded shaped product has a compressive strength of at least about 1200 psi.

48. The method of claim 44, wherein said shaping step comprises injection molding said product.

25 49. The method of claim 46, wherein the percent expansion of said expanded shaped product with respect to said product prior to said heating step is at least about 95%.

30 50. The method of claim 41, wherein said first thermoplastic resin is selected from the group consisting of the polystyrenes, solid rubbers, and mixtures thereof.

51. The method of claim 41, wherein said epoxy resin is a bisphenol A-based liquid epoxy resin.

35 52. The method of claim 42, wherein said second thermoplastic resin is selected from the group consisting of the polystyrenes, solid rubbers, and mixtures thereof.

59. The device of claim 58, said device including a plurality of said U-shaped resin bodies being positioned spaced apart from one another and in alignment with one another.

5 60. The device of claim 56, wherein said first thermoplastic resin is selected from the group consisting of the polystyrenes, solid rubbers, and mixtures thereof.

61. The device of claim 56, further including a quantity of a second thermoplastic resin different from said first thermoplastic resin and selected from the  
10 group consisting of the polystyrenes, rubbers, and mixtures thereof.

62. The device of claim 56, wherein said epoxy resin is a bisphenol A-based liquid epoxy resin.

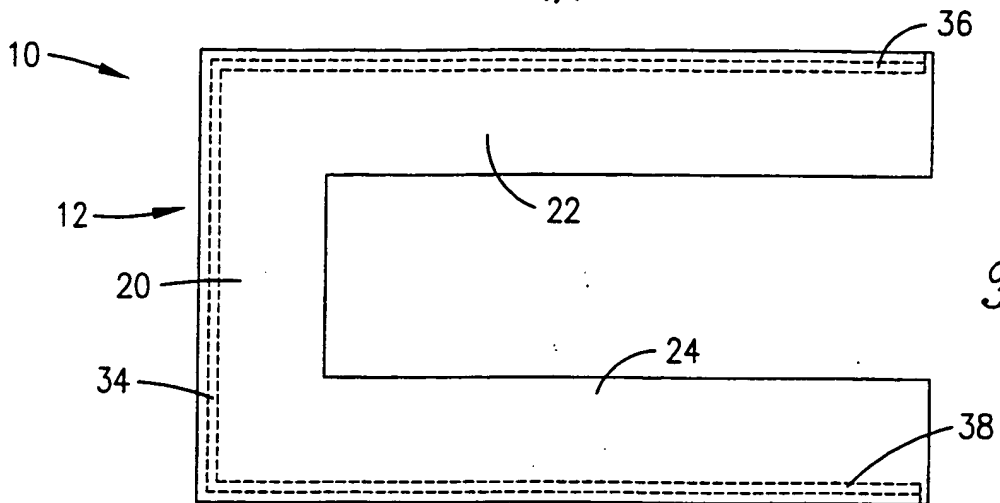
15 63. The device of claim 56, wherein said body is expanded by heating to a temperature of at least about 300°F and said expanded body has a compressive strength of at least about 1200 psi.

64. The device of claim 56, wherein said body is formed by injection  
20 molding.

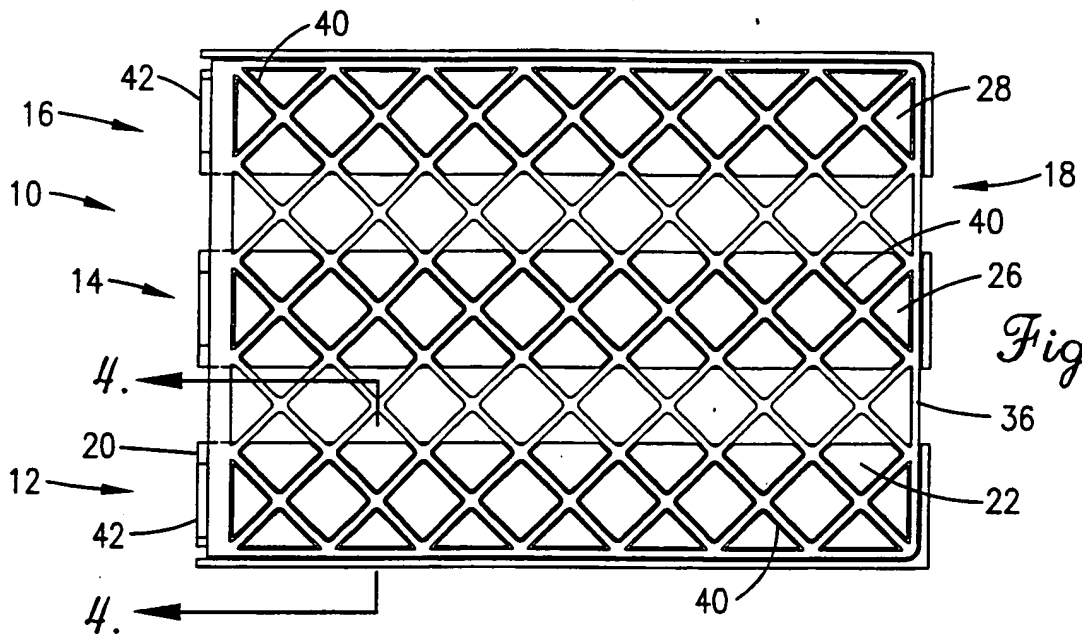
65. The device of claim 56, said body further including a compound selected from the group consisting of pigments, blowing agents, catalysts, curing agents, reinforcers, and mixtures thereof.

25 66. The device of claim 65, wherein said compound is a reinforcer selected from the group consisting of hydrated amorphous silica, glass microspheres, and mixtures thereof.

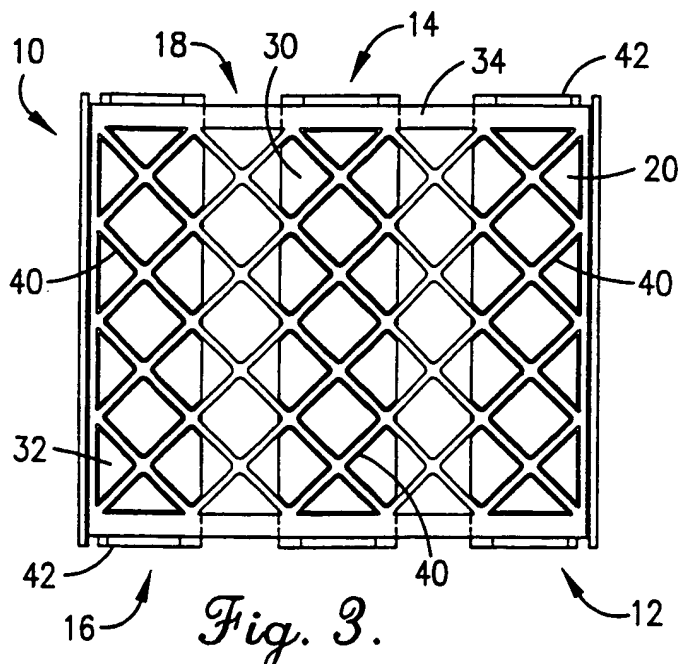
30 67. The device of claim 66, wherein said reinforcer is hydrated amorphous silica, and said hydrated amorphous silica is present in said body at a level of from about 1-10% by weight, based upon the total weight of the body taken as 100% by weight.



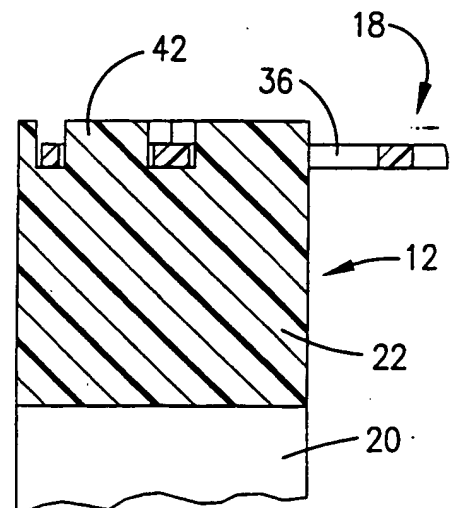
*Fig. 1.*



*Fig. 2.*



*Fig. 3.*



*Fig. 4.*

# INTERNATIONAL SEARCH REPORT

International application No.

PCT/JS99/24795

## A. CLASSIFICATION OF SUBJECT MATTER.

US CL. :

521/135; 428/83, 122, 158, 304.4; 264/45.4, 46.4, 46.9, 51, 140, 328.1



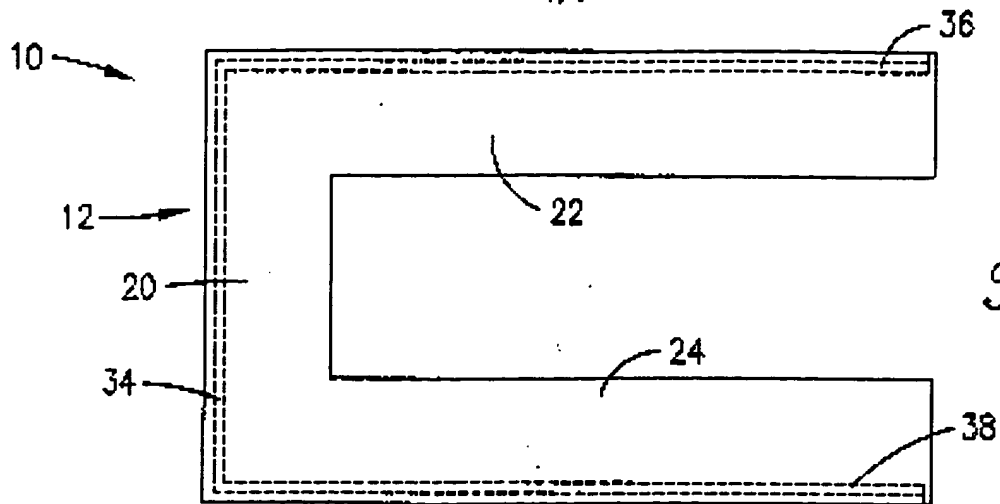


Fig. 1.

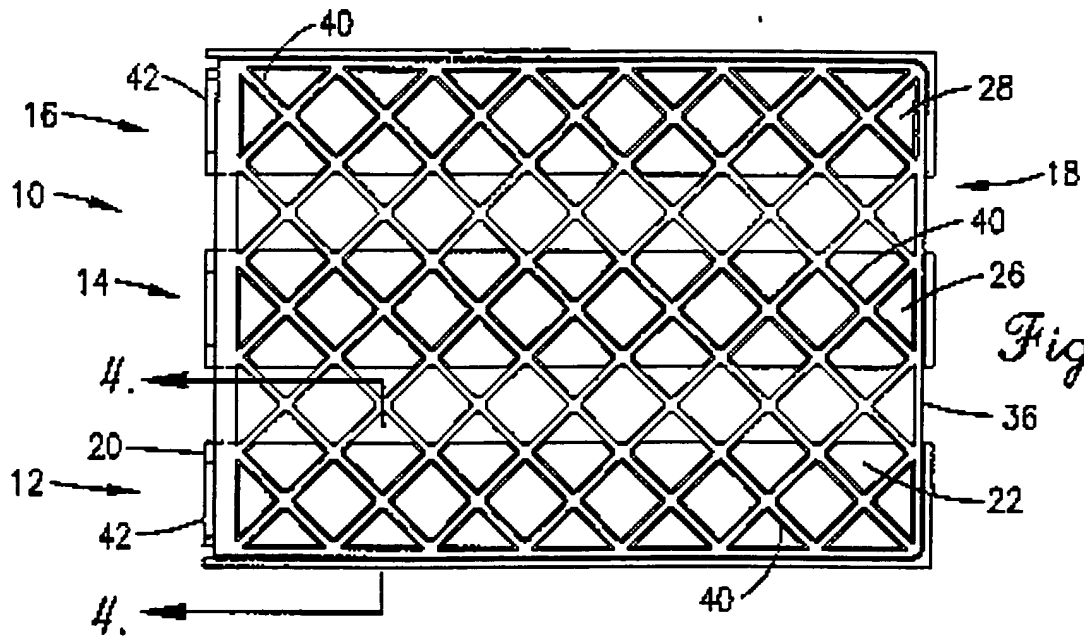


Fig. 2.

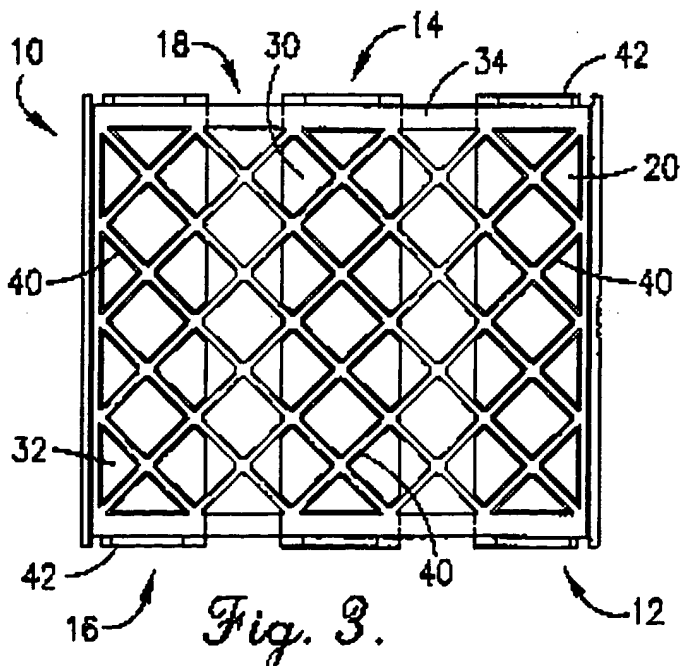


Fig. 3.

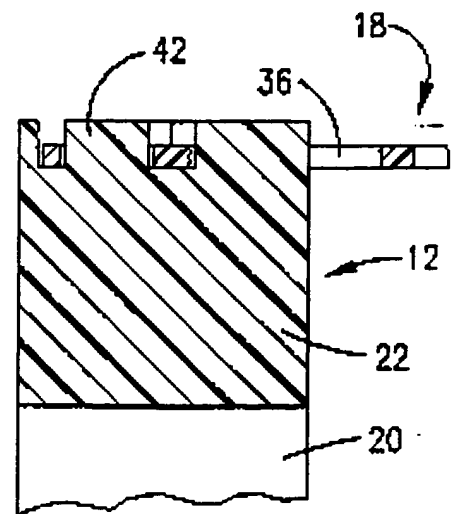


Fig. 4.

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